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Simulation of warpage induced by non-isothermal crystallization of co-Polypropylene during the SLS process

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Abstract. Polymer processing using Additive Manufacturing Technologies (AM) has experienced a remarkable growth during the last years. The application range has been expanding rapidly, particularly driven by the so-called consumer 3D printing sector. However, for applications demanding higher requirements in terms of thermo-mechanical properties and dimensional accuracy the long established AM technologies such as Selective Laser Sintering (SLS) do not depict a comparable development. The higher process complexity hinders the number of materials that can be currently processed and the interactions between the different physics involved have not been fully investigated. In case of thermoplastic materials the crystallization kinetics coupled to the shrinkage strain development strongly influences the stability of the process. Thus, the current investigation presents a transient Finite Element simulation of the warpage effect during the SLS process of a new developed polyolefin (co-polypropylene) coupling the thermal, mechanical and phase change equations that control the process. A thermal characterization of the material was performed by means of DSC, integrating the Nakamura model with the classical Hoffmann-Lauritzen theory. The viscoelastic behavior was measured using a plate-plate rheometer at different degrees of undercooling and a phase change-temperature superposition principle was implemented. Additionally, for validation porpoises the warpage development of the first sintered layers was captured employing an optical device. The simulation results depict a good agreement with experimental measurements of deformation, describing the high sensitivity of the geometrical accuracy of the sintered parts related to the processing conditions.

Keywords: Additive Manufacturing, Selective Laser Sintering, 3D-Printing, Polymer Crystallization, FEM Simulation
PACS: 44, 46, 81, 83

INTRODUCTION

Additive Manufacturing (AM) is currently a technology that is revolutionizing the way of how to produce parts or assemblies. Different technologies, gathered under the term 3D Printing, have entered the market for manufacturing objects ranging from simple visual prototypes till end use functional parts. AM presents several advantages in comparison to traditional manufacturing methods such as additional geometric complexity for free, shorter development cycle times and lower costs for small production series, but only some of them are able to produce functional parts [1]. Among these technologies, Selective Laser Sintering (SLS) is one of the most promising manufacturing techniques. However, the higher process complexity of this layer-wise production method hinders the number of materials that can be currently processed and the interactions between the different physics involved have not been fully investigated [2][3][4][5]. In this direction several efforts have been conducted to acquire a better understanding of the SLS process through process modelling. The initial model developed by Nelson et al. [6] employed a one dimensional finite element method to predict the density variations along the vertical build direction for an amorphous thermoplastic (polycarbonate). Ryder et al. [7] extended the previous model to 2 dimensions, incorporating also the effect of a spatially dependent conductivity and porosity. Other research groups have further studied other related phenomena such as Z growth [8], thermal degradation [9], polymer densification [10] and temperature evolution during laser scanning [11]. One of the main limitations for SLS polymer processability is related to the so-called warpage or curling effect that develops when the polymer starts crystallizing. However, only limited efforts have been conducted in this direction. A previous work performed by Jamal et al. [12] presents a simulation of the curling effect of a polycarbonate. However, nowadays the most used materials in the market correspond to semi-crystalline thermoplastics and the physics involved differ substantially due to the presence of a phase transition within processing. Therefore, the following investigation addresses for the first time this phenomenon for a semi-crystalline thermoplastic material employing a multi-physics Finite Element (FE) simulation approach.

THEORETICAL BACKGROUND

Crystallization Kinetics Theory

The overall crystallization kinetics theories describe the evolution of the relative degree of crystallization $\alpha(t)$ as a function of time t and temperature T. For this simulation the differential form of the model developed by Nakamura et al. [13] for a temperature dependent crystallization rate was considered:

$$\frac{\partial \alpha}{\partial t} = nK(T)(1-\alpha)\left[\ln\left(\frac{1}{1-\alpha}\right)\right]^{\frac{n-1}{n}} \tag{1}$$

where $K(T)$ is the non-isothermal crystallization rate. The Nakamura $K(T)$ and Avrami $k(T)$ rate constants can be related to each other by the following expression:

$$K(T) = k(T)^{\frac{1}{n}} = \ln(2)^{\frac{1}{n}}\left(\frac{1}{t_{1/2}}\right) \tag{2}$$

where $t_{1/2}$ is the corresponding half crystallization time for a defined isothermal temperature. According to Patel [14] the determination of the reciprocal half crystallization time can be expressed by the Hoffman-Lauritzen theory:

$$\left(\frac{1}{t_{1/2}}\right) = K_0 \exp\left(\frac{-U}{R(T-T_\infty)}\right)\exp\left(\frac{-K_G(T+T_o)}{2T^2\Delta T}\right) \tag{3}$$

where K_0 is a temperature independent constant, U is the activation energy of the crystallization transport which takes the universal value of 6270 J/mol, R is the universal gas constant equal to 8.314 J/mol/K and $T_\infty = T_G$-30°K is the temperature at which the crystallization transport finishes, with T_G the related glass transition temperature. K_G is related to the nucleation characteristics and $\Delta T = T_o - T$ corresponds to the under-cooling from the equilibrium melting point T_o, which can be determined by the Hoffman-Weeks construction [15]. To determine K_0, K_G and n different isothermal measurements need to be performed by means of differential scanning calorimetry (DSC).

Heat Transfer Model

The equation that governs the heat transfer process of the SLS sintered parts inside the build cylinder during the cooling stage is defined as follows:

$$\rho(\alpha,T)Cp(\alpha,T)\frac{\partial T}{\partial t} = \nabla\cdot\left(k(\alpha,T)\cdot\nabla T\right) + \rho(\alpha,T)\Delta H_c\frac{\partial \alpha}{\partial t} \tag{4}$$

where $\rho(\alpha,T)$ corresponds to the material density, $Cp(\alpha,T)$ to the material heat capacity and $k(\alpha,T)$ to the thermal conductivity. ΔH_c accounts for the crystallization enthalpy. The crystallization rate was previously defined in Equation 1. The material properties are also defined as a function of the relative crystallinity during the phase change and expressed by the following relations:

$$\begin{aligned} \rho(\alpha,T) &= \alpha\rho_s(T) + (1-\alpha)\rho_m(T) \\ Cp(\alpha,T) &= \alpha Cp_s(T) + (1-\alpha)Cp_m(T) \\ k(\alpha,T) &= \alpha k_s(T) + (1-\alpha)k_m(T) \end{aligned} \tag{5}$$

where the subscripts s and m denote respectively the solid and molten states of the bulk polymer.

Viscoelastic Mechanical Model

The Generalized Maxwell Viscoelastic model is considered for this study. The general equation that defines a linear viscoelastic shear modulus *G* with *p* spring and damper components (branches) is as follows:

$$G(t,\alpha,T) = G_\infty + \sum_{i=1}^{p} G_i(A_x(\alpha,T),t)$$

$$G_i(A_x(\alpha,T),t) = G_m(A_x(\alpha=1,T),T)\cdot\mu_i\cdot\exp\left(-\frac{t}{A_x(\alpha,T)\cdot\lambda_i}\right) \tag{6}$$

$$\mu_i = \frac{G_m(A_{xi}(\alpha,T),T)}{\sum_{i=1}^{p} G_m(A_{xi}(\alpha,T),T)}$$

The effective relaxation time constant of each branch $A_x\lambda_i$ is dependent on the crystallization degree α represented by the shift factor function A_x at a defined temperature *T* based on the model developed by Eom et al. [16]. In this case the contribution of each branch is related to the relative fraction of each unrelaxed shear modulus μ_i to the measured viscoelastic shear modulus mastercurve G_m of the fully crystallized material. In this study only the deviatoric strain is considered viscoelastic, while the volumetric strain remains elastic.

The general stress-strain equation results as follows:

$$\underline{\underline{\sigma}} - \underline{\underline{\sigma}}_0 = K\cdot trace\left(\underline{\underline{\varepsilon}} - \underline{\underline{\varepsilon}}_0 - \underline{\underline{\varepsilon}}_{inel}\right)\underline{\underline{I}} + 2\cdot G_\infty\left(\underline{\underline{\varepsilon}} - \underline{\underline{\varepsilon}}_0 - \frac{1}{3}trace\left(\underline{\underline{\varepsilon}} - \underline{\underline{\varepsilon}}_0\right)\underline{\underline{I}}\right) + \sum_{i=1}^{p} 2\cdot G_i\,\underline{\underline{q}}_i$$

$$\frac{\partial \underline{\underline{q}}_i}{\partial t} + \frac{1}{\tau_i}\underline{\underline{q}}_i = \frac{\partial}{\partial t}\left(\underline{\underline{\varepsilon}} - \underline{\underline{\varepsilon}}_0 - \frac{1}{3}trace\left(\underline{\underline{\varepsilon}} - \underline{\underline{\varepsilon}}_0\right)\underline{\underline{I}}\right) \tag{7}$$

where σ corresponds to the stress tensor, *K* to the bulk elastic modulus, ε to the strain matrix, and G_i to the viscoelastic shear moduli associated to a symmetric tensor q_i, where each component represents the extension of the corresponding spring on each branch. The inelastic strain corresponds to a combination of the linear bulk thermal expansion coefficient α_{linear} related to a reference temperature T_{ref} and to the bulk volume contraction during the phase change $\Delta V_{shrinkage}$ due to the crystallization effect as defined in Equation 8.

$$\underline{\underline{\varepsilon}}_{inel} = \alpha_{linear}\left(T - T_{ref}\right)\underline{\underline{I}} + \left(1 - \left(1 - \Delta V_{shrinkage}\right)^{\frac{1}{3}}\right)\alpha\underline{\underline{I}} \tag{8}$$

EXPERIMENTS AND SIMULATION RESULTS

Materials Characterization Data

For this study a new SLS thermoplastic polymer was considered known as icoPP (Inspire irpd, Switzerland). This material corresponds to a co-polypropylene produced by a co-extrusion method. The crystallization constants were obtained employing DSC measurements at different isothermal/non-isothermal temperatures/rates between 106ºC and 110ºC obtaining values of n=2.44, K_0=3.924e10 1/s and K_G=498964 K^2. The linear expansion coefficient α_{linear}=254e-6 1/K and phase change volume contraction $\Delta V_{shrinkage}$=0.09 were obtained from [17]. More details about the thermal characterization set up can be found in the previous work [18]. The viscoelastic characterization measurements were performed using a plate-plate (ϕ 25 mm) M301 Anton Paar rheometer using 4 isothermal testing conditions. Each experiment considers a continuous frequency sweep while crystallization develops at undercooling temperatures of 106ºC, 108ºC, 110ºC and 112ºC with a sweep cycle period of 1 min. The frequency response was measured at 7 points per decade between 0.1 Hz and 10 Hz with strain amplitude of 0.1%.

Measurement of Deformation & Simulation Results

Figure 1 presents the experimental configuration to characterize the transient out of plane deformation of a 3 layer sintered disc in a SLS equipment and the model used for the FE simulation.

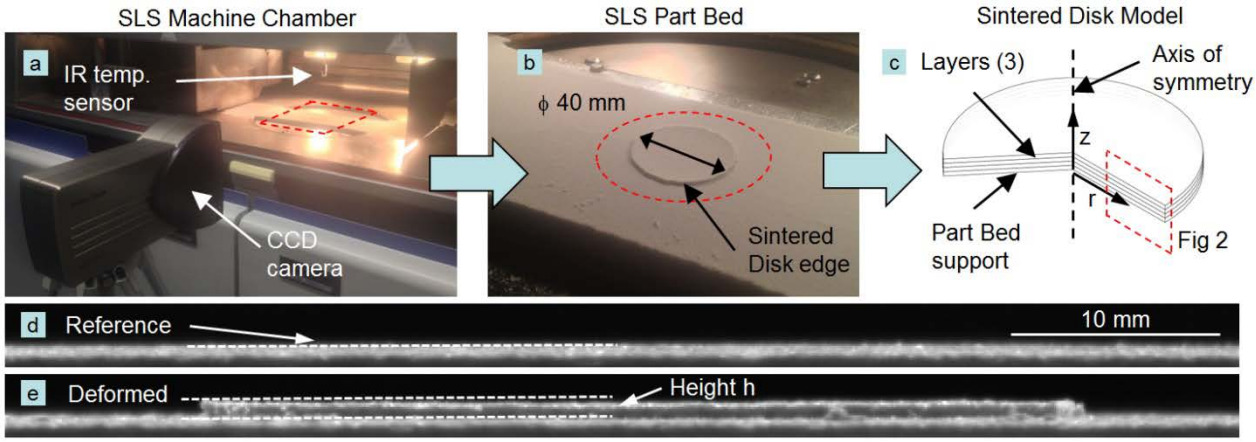

FIGURE 1. Experimental set up for the measurement of out of plane deformation of a 3 layer sintered disk

As observed in Figure 1(a) a CCD camera was placed in front of the SLS machine chamber at the same level of the part bed surface. For this configuration an 8 bit gray scale image with a resolution of 96 mm/pixel was obtained with a frame capture rate of 3 images per second. The lateral view of the disk is depicted in Figure 1(d), corresponding to the reference condition, where warpage has not been developed. Image 1(e) depicts the deformed configuration once the disk is fully crystallized and the height *h* reached a maximum stable value. An infrared temperature sensor records simultaneously the temperature evolution of the top surface of the sintered disk. Figure 1(c) presents the FE geometry used for the simulation (3 circular layers with diameter of 40 mm). In the axisymmetric simulation the transient addition of layers is considered. After the last layer of powder is deposited the chamber cools down from the T_{ref}=114ºC and the measured temperature is used as a boundary condition for the FE model. The bottom of the layer is considered thermally isolated and a non-penetration boundary condition is used as vertical support for the stack of layers.

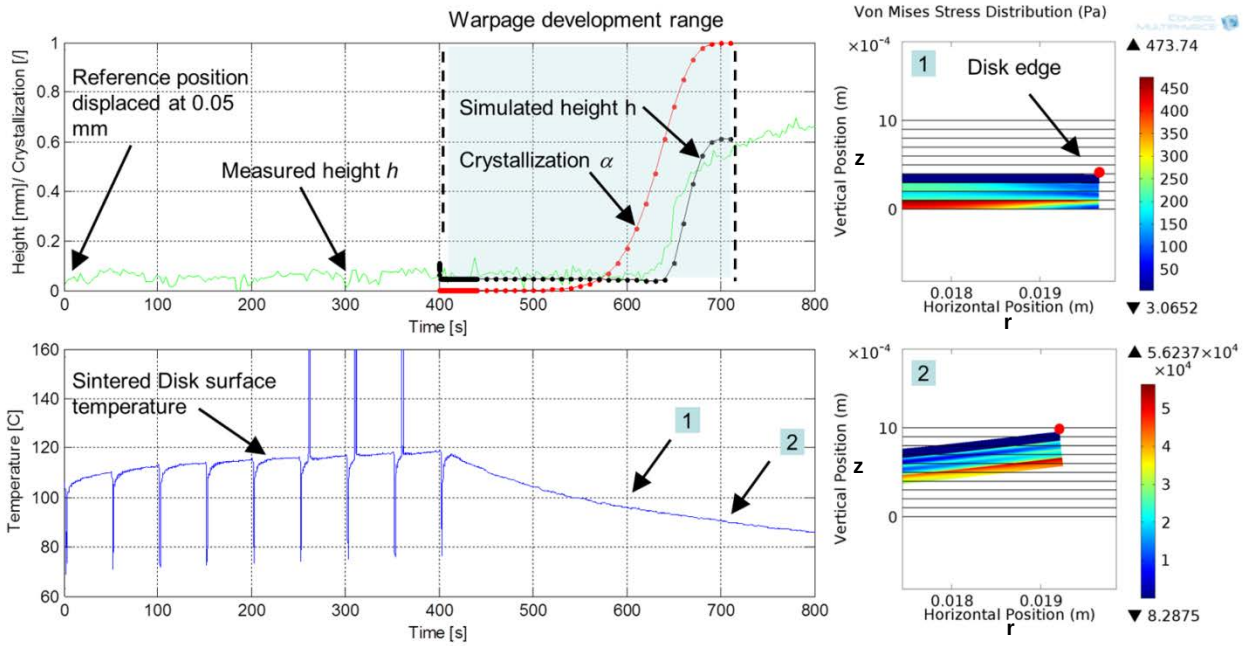

FIGURE 2. Experimental and simulated out of plane deformation in correlation with the disk transient temperature profile

Figure 2 presents the results for the measured and simulated height *h* and the evolution of the crystallization degree α at a point on the disk edge. As observed, the simulation results predict accurately the initiation of warpage at 640s. At this time, the crystallization degree is above 50%, which indicates that early stages of crystallization for this material do not influence the warpage development. Additionally the stress distribution observed at the disk cross section results highly inhomogeneous, particularly between different layers. This behavior is associated to the different crystallization degree along the build direction as reported in a previous work [18]. Before the out of plane deformation starts to develop, the disk presents a maximum equivalent stress of 473 Pa at the bottom (1st layer). At the end of the crystallization phase the maximum value reaches 56237 Pa, which results more than 100x higher than the initial condition, which depicts that residual stress distributions can highly influence the final geometric accuracy of SLS sintered parts.

SUMMARY AND OUTLOOK

The present work presented a new model to describe the warpage development during the crystallization phase of a thermoplastic material during SLS processing. For the first time a multiphysics approach was used to connect the thermal, mechanical and phase change physics in order to quantitatively describe this phenomenon. A suitable correlation between simulation and experimental results was obtained for this particular configuration. Such a model helps to better understand the stress-strain development during the process and could be used in the future to predict geometric inaccuracies and optimize the location and orientation of complex shapes within the building chamber.

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